

PATENT SPECIFICATION

DRAWINGS ATTACHED

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COMPLETE SPECIFICATION

Manufacture of small spherical particles

We, SOUTHWEST RESEARCH INSTITUTE, of 8500 Culebra Road, San Antonio, Texas, United States of America, a corporation organised and existing under the laws of the State of Texas, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates generally to a method and apparatus for manufacturing small spherical particles, and has special (but not exclusively) utility in the manufacture of such particles in the form of capsules in which a filler material is contained within a seamless film material. More particularly, this invention relates to improved techniques for producing such particles, and especially such capsules, on a mass production basis.

In accordance with one prior technique, capsules of the type described are mass produced by means of centrifugal action. Thus, as shown and described in my prior U.S. patent No. 3,015,128, a liquid film material is supplied to the interior of a rotating drum so as to extend across the inner ends of a bank of orifices disposed about and extending through the outer wall of the drum. Globules of the filler material are then delivered to the film material across the orifices, and the speed of rotation of the drum is such that the resulting centrifugal force overcomes the adhesive forces of the film material at the orifices so that it forms a shell about each filler material globule and is severed and flung outwardly from the orifices. As each capsule is so severed, additional film material forms across the orifice to receive an additional filler material globule, and all the fluid capsules are hardened in a suitable manner, such as within a bath

of liquid hardening medium located to receive the capsules as they are flung from the orifices of the encapsulating drum.

Although this represents a substantial advance in the encapsulating art, it nevertheless has certain shortcomings when the filler material and film material have appreciably different densities. Thus, in this event, the centrifuging effect has a tendency to displace the globule of filler material off-center with respect to the shell of liquid film material. This eccentricity may be aggravated when the fluid capsules are flung into a liquid hardening medium, because of the impact of the capsule with the medium or collision with another capsule.

In accordance with another prior technique, the filler and film materials are extruded in the form of concentric fluid rods, which are caused to break off into individual fluid capsules by gravity or by vibration. Although this method is not as sensitive to density differences between the filler and film materials at the centrifugal method, it is nevertheless a much slower process and thus not capable of as high a rate of production. Furthermore, when the fluid capsules are hardened in a liquid hardening medium, there is the additional problem, above mentioned, of shifting the filler material off-center due to impact of the capsules with the medium as well as collision with one another. These latter problems increase as the extrusion rate is increased, because of the more severe impact of the fluid capsules with the fluid hardening medium.

A still further difficulty encountered with the use of a liquid hardening medium, in these or other methods of forming spherical particles in which successive particles are directed toward the same point in the hardening medium, is their tendency to agglomerate.

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There is often a need, in different types of known encapsulating methods, for accurately controlling the temperature of the fluid capsule when the film material is a hot melt.

5 Shells of the film material are frequently of low strength when in the fluid state, and the rate at which they are cooled is critical. Solidification at too rapid a rate may result in deformed capsules, whereas solidification at too low a rate may result in excessive capsule breakage.

10 An object of this invention is to provide a method and apparatus for mass producing spherical particles, whether in solid or capsule form, which obviates one or more of the foregoing difficulties, while at the same time enabling a very high rate of production.

15 The invention provides a method of mass producing small spherical particles, comprising the steps of extruding a rod of liquid material within a confined stream of carrier fluid which is chemically nonreactive and physically immiscible with said material and which is flowing at a speed greater than the rate of extrusion of said rod and moving in the same direction as said rod so as to cause it to elongate and break off into segments which form individual spherical particles, 20 and then hardening said spherical particles.

25 The method according to the invention preferably includes the further step of extruding a second rod of liquid material concentrically within the first-mentioned rod, so as to form a composite rod which is caused to elongate and break off into segments which form capsules in which the segments of said first rod form a seamless film about the segments of said second rod.

30 The invention also provides apparatus for use in mass producing small hardened seamless capsules, comprising a confined conduit, a pair of concentric tubes in the conduit, means for feeding liquid filler and film material into the inner and outer tubes, respectively, so as to extrude concentric fluid rods of said materials into said conduit, and means for forcing a stream of fluid carrier through the conduit and about the end of the tubes therein in the same direction as the extrusion and at a greater speed than the rate of extrusion of said materials so as to cause the extruded concentric rods to elongate and break off into segments which form individual seamless capsules of filler material within said film material.

35 The term "confined stream" means that the stream is enclosed, as in a pipe, and not open, as in a trough or flume. The term "confined conduit" means a closed conduit such as a pipe and is intended to exclude open conduits such as a trough or flume.

40 The stream of carrier fluid is influential in separating the particles to minimize the possibility of collision prior to hardening.

The size of the particles is inversely proportional to the speed of the carrier fluid, so that the latter may be regulated to form particles of different sizes.

45 Since this invention involves neither a centrifuging action nor any other action tending to move the filler material off-center, the resulting capsule will have a shell of uniform thickness.

50 In one embodiment of the invention, the shell of film material comprises a hot melt which is hardened while suspended in the stream of carrier fluid. Thus, the carrier fluid is maintained at a temperature above the melting point of the film material as it flows past the extruded rod and then reduced below such point to harden the fluid capsules. As a result, the capsules are hardened before colliding with one another upon separation from the carrier fluid and collection. Also, of course, the heating of the carrier fluid permits the maintenance of a desired temperature gradient for the capsules.

55 In another embodiment, the shell is instead a material which is hardened by chemical reaction or by solvent extraction. In this case, the fluid capsules are introduced with the carrier fluid into a suitable liquid hardening medium. However, the carrier fluid in which the capsules are suspended will distribute them to random locations within the medium. Also, the stream of carrier fluid will agitate the medium to a certain extent, so as to further reduce the likelihood of the capsules colliding with one another before hardening in the medium. Still further, the carrier fluid is a liquid, such as water, collected in a vessel above the hardening medium contained therein, so that the fluid capsules settle gradually to random locations of the medium, thereby essentially eliminating impact.

60 The stream of carrier fluid is confined within a conduit connecting at its downstream end with the vessel for collecting the carrier fluid and separating the hardened capsules therefrom. The upstream end of the conduit is connected with a suitable means for pumping the carrier fluid there-through at the desired speed. Preferably, the carrier fluid is recirculated from the vessel to the upstream end of the conduit by means of an auxiliary conduit connecting the vessel with the pumping means.

65 The concentric rods of fluid filler and film material are extruded from the ends of concentric tubes which extend into the conduit. Preferably, these tube ends are arranged concentrically within the conduit and face toward its downstream end.

70 The accompanying drawings show, by way of illustration, the above-mentioned two embodiments of the invention:

Fig. 1 is a diagrammatic view of appa- 130

tus constructed in accordance with the first-mentioned embodiment of this invention; and

Fig. 2 is a diagrammatic view of apparatus constructed in accordance with the second-mentioned embodiment of the invention.

With reference now to the details of the above-described drawings, the first-mentioned embodiment of the apparatus shown in Fig. 1 comprises a conduit 10 having an upstream section 10a of enlarged circular cross-section and a downstream section 10b of reduced circular cross-section and connected to the upstream section by means of a conical restriction 10c. Inner and outer concentric tubes 11 and 12 extend concentrically within enlarged conduit section 10a for extruding fluid rods of filler and film material into the conduit. As shown in Fig. 1, the open ends of the tubes 11 and 12 terminate substantially adjacent the conduit restriction 10c, while the opposite ends of the tubes extend through the closed end 13 of conduit section 10a for connection with reservoirs 14 and 15. Thus, film material may be contained within reservoir 14 for supplying it to the outer tube 12, and filler material may be contained within a reservoir 15 for supplying it to the inner tube 11. These materials are kept in a fluid state as they are forced from the reservoirs, through pipes 16 and 17 and into the tubes by means of metering pumps 18.

Carrier fluid is forced into the upstream end of enlarged conduit section 10a so as to flow concentrically about the outer tube 12, and past the ends of the tubes, through the restriction 10c, and into the reduced section 10b of the conduit. Obviously, as this carrier fluid flows through the restriction 10c, its velocity is increased, depending upon the relative sizes of the annular flowway within the section 10a and the flowway within section 10b. The velocity is increased at this area adjacent the ends of the tubes 11 and 12 to a value sufficiently greater than the extrusion velocity of the film and filler materials through these tubes as to cause the concentric rods extruded therefrom to break off and form fluid capsules. As these capsules are broken off from the rods, they will flow suspended within the stream of carrier fluid through the reduced conduit section 10b.

As previously described, in this first embodiment of the invention, the film material which forms the shell of the capsule is a hot melt which is adapted to be hardened when cooled below its melting point. Thus, the stream of carrier fluid about the tubes is maintained at a temperature above the melting point of the film and filler material so that the fluid rods will remain liquid as they are extruded from the ends of the tubes

and caused to break off and form into fluid capsules. These capsules then flow suspended within the stream of carrier fluid into and through the reduced conduit section 10b. As shown in Fig. 1, there is a counterflow heat exchanger 19 disposed about reduced section 10b for gradually decreasing the temperature of the carrier fluid from a point above the melting point of the film material to a point below such point. Thus, the suspended fluid capsules are caused to harden during their flow from the upstream to the downstream end of the reduced conduit section 10b. More particularly, the temperature gradient is in this way controlled during this time so as to avoid damage to the shell material, as might incur in the event the temperature of the film material was too rapidly or too slowly reduced.

Carrier fluid is supplied to the upstream end of enlarged conduit section 10a by means of an auxiliary conduit 20 connected at its upstream end with a source of carrier fluid, to be described hereinafter, and at its downstream end with the upstream end of the enlarged conduit section. This carrier fluid is moved through the auxiliary conduit and into the main conduit 10 at the desired speed by means of a pump 21 disposed within the auxiliary conduit upstream of a flowmeter 22 for indicating the flow rate of the carrier fluid. The temperature of this carrier fluid is raised to the desired level by means of a heat exchanger 23 in the auxiliary conduit between the flowmeter 22 and the upstream of reduced conduit section 10a.

The downstream end of reduced conduit section 10b is connected with a vessel 24 for collecting the carrier fluid and separating the capsules suspended therefrom. In this embodiment of the invention, it is contemplated that the carrier fluid will be a liquid, such as water, of heavier specific gravity than the capsules. Thus, there is an outlet 25 from the vessel above the connection thereto of conduit section 10b, so that the hardened capsules rise to the upper level of the carrier fluid so as to spill with it over the edge of outlet 25 onto a screen conveyor 26. The hardened capsules will move on the conveyor from left to right so as to spill into a collection container 27, and a brush 28 is mounted beneath the right-hand end of the screen 26 for wiping capsules from the screen which tend to stick to it.

As previously described, the carrier fluid is preferably recirculated from the vessel 24 to the upstream end of the main conduit 10. For this purpose a container 29 is mounted beneath the left-hand end of the screen 26 to receive the carrier fluid which passes through it. Additional carrier fluid for make-up purposes may be introduced into the container 29 through a line 30 connect-

ing therewith. The lower end of carrier fluid container 29 is connected with the upstream end of auxiliary conduit 20, so that carrier fluid may be recirculated into the main conduit 10. This recirculation of carrier fluid has several advantages, particularly when it is of a relatively valuable material. Even when it is of relatively expendable material, its recirculation permits the retention of some of its heat used in maintaining the film material in a fluid state within at least a portion of conduit 10.

One or both of the tubes 11 and 12 may be adjusted longitudinally of the conduit 10, as well as with respect to one another in any conventional manner. In being adjustable relative to the conduit 10, the open ends of such tubes may be moved toward or away from the restriction 10c for adjusting the velocity at which the stream of carrier fluid moves about the extruded rods. The adjustment of the ends of the tubes relative to one another may be desirable in order to accomplish certain extrusion characteristics.

As previously described, the carrier fluid is chemically nonreactive and physically immiscible with the film material which forms the shell of the capsule. At the same time, the film material is a hot melt which may be hardened in response to the lowering of the temperature of the carrier fluid from a point above to a point below its melting point. Still further, the filler material is a substance which may be maintained in its fluid state at a temperature at which the film material is also maintained in its fluid state. Obviously, however, the filler material may or may not be hardened after extrusion. As an example of materials meeting these qualifications, the film material may comprise a molten wax, the carrier fluid may be water, and the filler material may also be water. These substances were formed into capsules approximately 1,000 microns in diameter and containing approximately 57% water with apparatus having the following structural and operation characteristics:

50 STRUCTURAL CHARACTERISTICS

Enlarged conduit section	0.580" I.D.
Reduced conduit section	0.315" I.D.
Outer tube	0.1875" O.D.
	0.150" I.D.
55 Inner tube	0.050" O.D.
	0.030" I.D.

Outer ends of the inner and outer tubes flush with one another and located $1\frac{1}{2}$ " upstream of the conical restriction.

60 OPERATIONAL CHARACTERISTICS

Upper Carrier Fluid Temperature	80° C.
Lower Carrier Fluid Temperature	50° C.

Filler Material Extrusion Rate	30 cc/min	65
Encapsulating Medium Extrusion Rate	30 cc/min	
Carrier Fluid Rate	$1\frac{1}{2}$ gpm	
Filler Material Temperature	80° C.	70
Encapsulating Medium Temperature	80° C.	

The same film, filler and carrier fluid materials were used in forming capsules approximately 1500 microns in diameter and containing approximately 57% water employing the same apparatus and the following operational characteristics:

Upper Carrier Fluid Temperature	80° C.	80
Lower Carrier Fluid Temperature	45° C.	
Filler Material Extrusion Rate	60 cc/min	
Encapsulating Medium Extrusion Rate	60 cc/min	85
Carrier Fluid Rate	2 gpm	
Filler Material Temperature	85° C.	
Encapsulating Medium Temperature	85° C.	

The other embodiment of the apparatus shown in Fig. 2 is similar, in many respects, to the apparatus above-described in connection with Fig. 1. Thus, it includes a conduit 10 having enlarged and reduced cylindrical sections 10a and 10b connected by a restricted portion 10c, and concentric tubes 11 and 12 extending concentrically within enlarged conduit section 10a. As in the first-described apparatus, filler material is fed in a liquid state to inner tube 11 and film material is fed in a liquid state to outer tube 12 from supply reservoirs 15 and 14, respectively, through pipes 17 and 16 having pumps 18 disposed therein. Also, carrier fluid is introduced into the upstream end of enlarged conduit section 10a at the desired speed through an auxiliary conduit 20 connected at its opposite end to a source of supply of carrier fluid. As in the first embodiment of the invention, such carrier fluid is forced through the auxiliary conduit by means of a pump 21, and the flow rate thereof is measured by means of a meter 22.

However, as previously mentioned, in this second embodiment of the invention, the fluid capsules broken off from the fluid rods extruded through the tubes 11 and 12 are hardened either by chemical reaction or solvent extraction, so there is normally no need for heat exchangers either in the restricted conduit section 10b or in the auxiliary conduit 20. Thus, in this second embodiment, the suspended fluid capsules flow entirely through the restricted conduit 10b in a fluid state and into a vessel 31 adapted to contain a liquid hardening medium as well as to collect the carrier fluid and capsules.

In the illustrated embodiment of Fig. 2,

it is contemplated that the carrier fluid will be a liquid having a lower specific gravity than the liquid hardening medium, and that the fluid capsules will have a specific gravity intermediate that of the fluid carrier and hardening medium. Furthermore, the liquid carrier fluid and hardening medium are also immiscible with one another so that they will stratify in the vessel to establish an interface 33. The end of the restricted conduit section 10b turns down to a level within the vessel below the upper level 32 of the carrier fluid, but above the interface 33 between the carrier fluid and hardening medium. As a result, the liquid capsules will gradually fall through the carrier fluid to the interface level 33. Also, this introduction of carrier fluid into the vessel 31 will agitate the fluid within the vessel so as to normally distribute the fluid capsules thereabout, and thus reduce the possibility of collision with one another prior to hardening at the interface level 33.

The interface level 33 is maintained by level control 34 at the upper open end of overflow tube 35. The lower end of tube 35 is disposed above a screen conveyor 36 similar to the screen conveyor 26 shown in Fig. 1. Thus, the hardening fluid capsules and hardening medium spill over the open upper end of the tube onto the screen conveyor 36, where, as described in connection with the apparatus of Fig. 1, the capsules are moved by the conveyor from left to right so as to spill into a container 37 disposed beneath the right-hand end of the conveyor. Also, blade 38 is used to wipe capsules from the conveyor in the event they have a tendency to stick to it.

The liquid hardening medium which spills over into the tube 35 and through the screen 36 is collected within another container 39 having its lower end connected by a pipe 40 to a pump 41, which recirculates it through a pipe 42 into the lower end of the vessel 31 beneath the interface 33. In the event there is a loss of hardening medium, as a result of which hardening medium no longer spills into tube 35, the level control 34 will, through suitable means indicated by broken lines in Fig. 2, open a valve 43 within a make-up line 44 for admitting additional liquid hardening medium into the pipe 40 and thus into the lower end of the vessel 31. When the interface level has thus been brought back up to the desired level, the level control 34 shuts the valve in the make-up line.

The upstream end of auxiliary conduit 20 is connected to the vessel 31 above the interface 33, but below the upper level 32 of the carrier fluid, so that carrier fluid will flow into the conduit 20 and thus to the pump 21 and through flow meter 22 for recirculation into the upstream end of the

main conduit 10. There may be need for a scrubber 45 in the auxiliary conduit in order to assure that there is no premature hardening of the film material within the main conduit 10.

As an example, this second embodiment of the invention may be used in the mass production of capsules comprising a filler material of hexane enclosed within a shell of aqueous sodium alginate. In this case, the carrier fluid may be naphtha, and the hardening medium may be aqueous calcium chloride, which converts the sodium alginate to insoluble calcium alginate. In this event, the carrier fluid might be scrubbed by liquid-liquid contact with water in the scrubber 45.

This invention has been illustrated and described in connection with the mass production of seamless capsules, because, as previously noted, it is especially well suited to such use. However, as also previously described, it also has utility in the mass production of solid spherical particles, and the term "particles", as used in the claims, contemplates either solid particles or particles in capsule form. As an example, it is contemplated that with suitable modification of the extruding means, this apparatus could be used in producing solid particles of wax. In this case, the carrier fluid could be water, which is immiscible with the wax and which may be maintained at a temperature level above and then lowered to a temperature level beneath the melting point of the wax as the fluid rod of the wax is broken off by means of the stream of carrier fluid.

From the foregoing, it will be seen that this invention is one well adapted to attain all of the ends and objects hereinabove set forth, together with other advantages which are obvious and which are inherent to the apparatus.

WHAT WE CLAIM IS:—

1. A method of mass producing small spherical particles, comprising the steps of extruding a rod of liquid material within a confined stream of carrier fluid which is chemically nonreactive and physically immiscible with said material and which is flowing at a speed greater than the rate of extrusion of said rod and moving in the same direction as said rod so as to cause it to elongate and break off into segments which form individual spherical particles, and then hardening said spherical particles.
2. A method as defined in Claim 1, including the further step of extruding a second rod of liquid material concentrically within the first-mentioned rod, so as to form a composite rod which is caused to elongate and break off into segments which form capsules in which the segments of said first

rod form a seamless film about the segments of said second rod.

3. A method as defined in Claim 1 or Claim 2, including the step of maintaining the carrier fluid at a temperature above the melting point of the liquid particles or the capsules as it flows past the first rod or the composite rod and then reducing the temperature of the carrier fluid below such point to harden the particles or the capsules while suspended in said stream of carrier fluid.

4. A method as defined in Claim 1 or Claim 2, including the step of introducing the carrier fluid and the liquid particles or capsules suspended therein into a liquid medium so as to harden the particles or capsules.

5. A method as defined in any one of Claims 1 to 4, wherein the confined stream of carrier fluid is forced concentrically about the end of an orifice from which the first rod or composite rod is extruded.

6. Apparatus for use in mass producing small hardened seamless capsules, comprising a confined conduit, a pair of concentric tubes in the conduit, means for feeding liquid filler and film material into the inner and outer tubes, respectively, so as to extrude concentric fluid rods of said materials into said conduit, and means for forcing a stream of fluid carrier through the conduit and about the end of the tubes therein in the same direction as the extrusion and at a greater speed than the rate of extrusion of said materials so as to cause the extruded concentric rods to elongate and break off into segments which form individual seamless capsules of filler material within said film material.

7. Apparatus as defined in Claim 6,

wherein the inner diameter of said conduit is reduced adjacent the ends of the tubes so as to increase the velocity of said carrier fluid.

8. Apparatus as defined in Claim 7, including means for adjusting the position of the tubes longitudinally of the conduit.

9. Apparatus as defined in any one of Claims 6 to 8, including a vessel at the end of the conduit downstream of the tubes for collecting the fluid carrier and capsules suspended therein, means for separating the hardened capsules from the fluid carrier, and means for recirculating the fluid carrier from the vessel to the end of the conduit upstream of the ends of the tubes.

10. Apparatus as defined in Claim 9, including means for cooling the fluid carrier within the conduit between the ends of the tubes and the vessel.

11. Apparatus as defined in Claim 9, including means for introducing a liquid hardening medium into said vessel.

12. The method of forming capsules hereinbefore described with reference to Figure 1 of the accompanying drawings.

13. The method of forming capsules hereinbefore described with reference to Figure 2 of the accompanying drawings.

14. The apparatus for forming capsules hereinbefore described with reference to Figure 1 of the accompanying drawings.

15. The apparatus for forming capsules hereinbefore described with reference to Figure 2 of the accompanying drawings.

TREGGAR, THIEMANN & BLEACH,

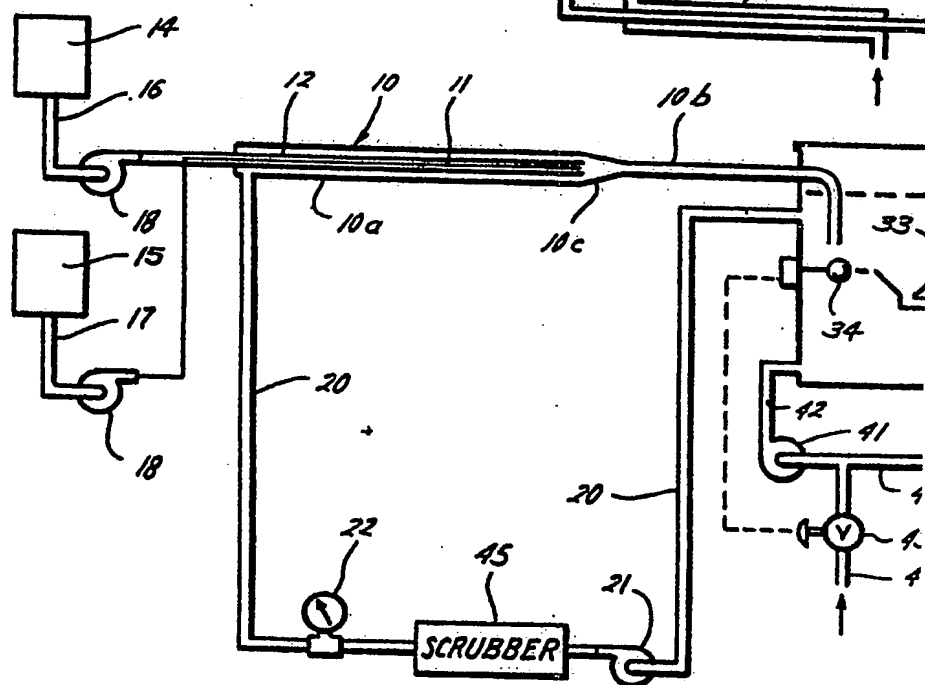
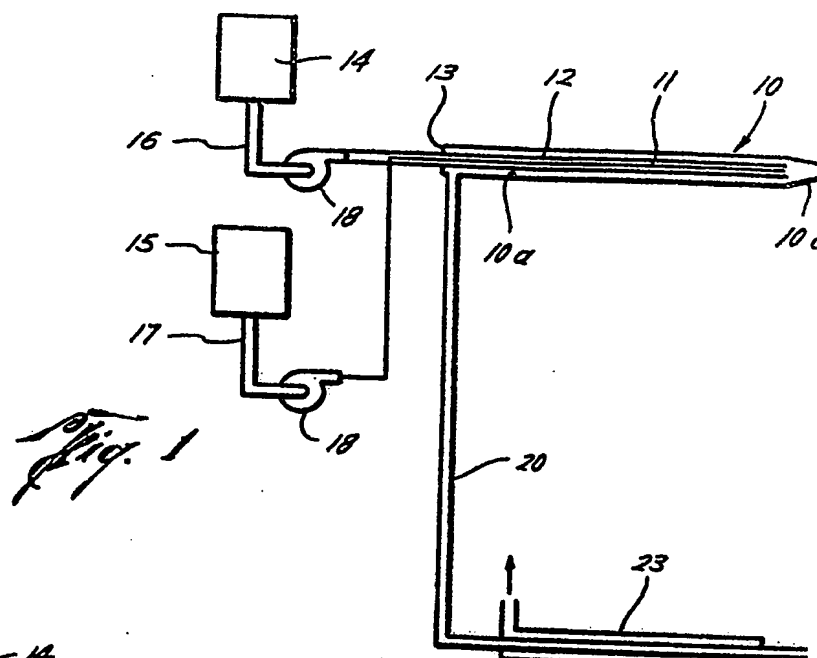
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COMPLETE SPECIFICATION

1 SHEET

This drawing is a reproduction of the Original on a reduced scale.

